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Aluminum Release Isentropes from Quantum Molecular Dynamics Simulations

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U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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Computational Project for a Theorist

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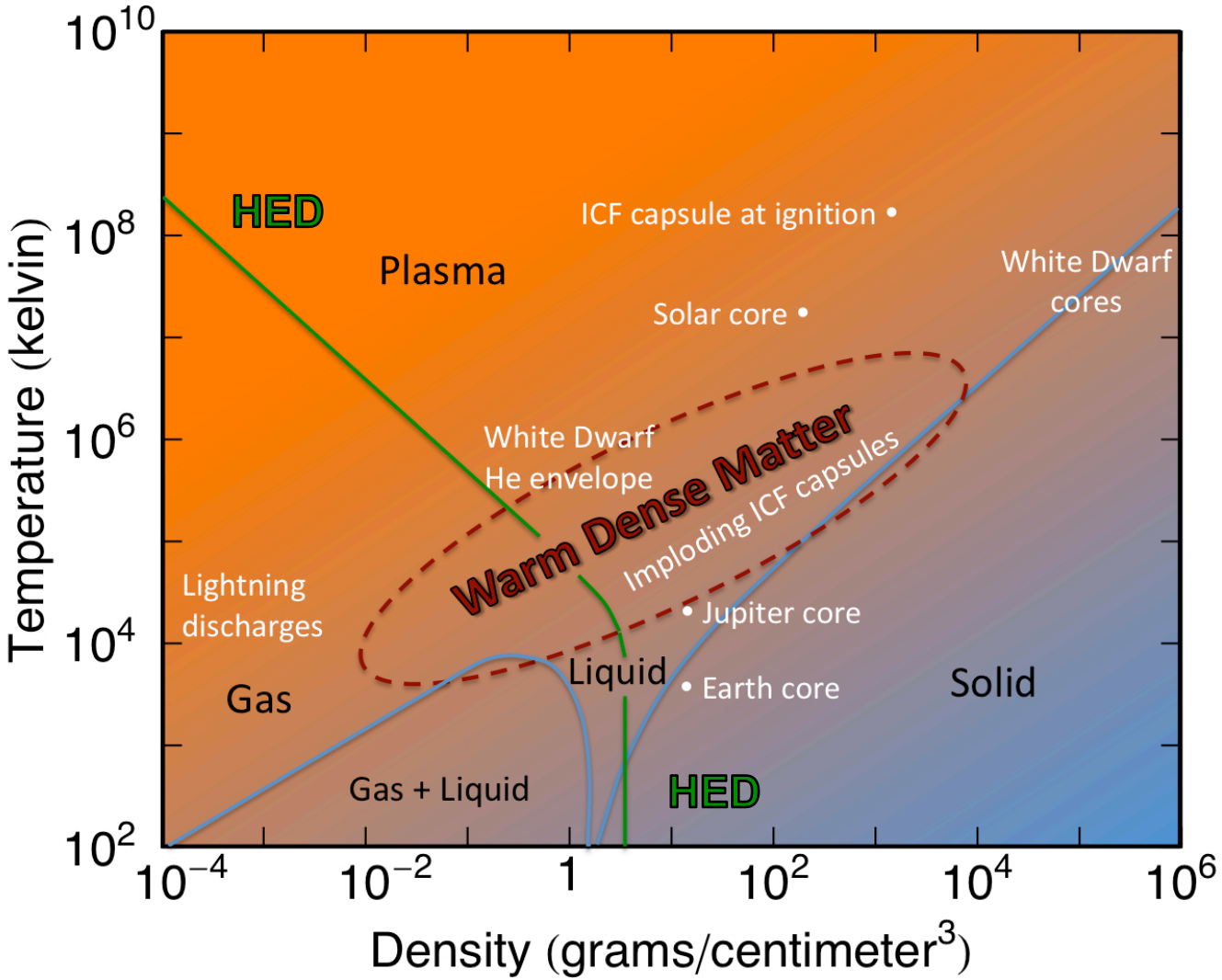
- Three-month practicum as component of Department of Energy Computational Science Graduate Fellowship
- Supervised by Mike Desjarlais and trained by Kyle Cochrane
- Preparation: Computational Methods in High Energy Density Plasmas long program at UCLA's Institute for Pure and Applied Mathematics
- Project Goals
  - **Compare two methods for computing release isentropes for aluminum**
  - Gain basic skills with VASP
  - Increase knowledge of shock physics
  - Become familiar with life at the Labs
  - Learn how my thesis results might affect real calculations

Warm Dense Matter: Quantum or Classical?



Models and Predictions Rely on Equations of State

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- Engineering codes demand knowledge of material properties over entire range of interest
- Historical models fail for warm dense matter (WDM)
- Different EOS and databases give different material properties
- Experimental data is limited to isolated regions of phase space
- Massive databases require reliable computational methods
  - Must incorporate quantum and classical behavior
  - Prediction of quality and size of errors is ideal
  - Good agreement with available experimental data

VASP: Quantum Molecular Dynamics (QMD) for WDM

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- Vienna Ab-Initio Software Package (VASP): combines quantum treatment of electrons with classical ions
- Density functional theory: highly efficient quantum method
- Molecular Dynamics: classical mechanics for simulating ion lattices
- QMD output can be used to constrain EOS parameters
- Shown to give good agreement with experimental data
  - Example: M. P. Desjarlais Phys. Rev. B **68**, 064204 (2003)

Reliable Hugoniot Calculations Using VASP

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- Materials under shock compression satisfy the Rankine-Hugoniot relation:

$$2\left(E_2 - E_1\right) = \left(P_2 + P_1\right)\left(\upsilon_1 - \upsilon_2\right)$$

where 1 and 2 are initial and final states, E is energy, P is pressure, and v is specific volume.

- Relation derived from conservation of mass, energy, and momentum

- Two methods: ramps and bracketing
  - Ramps: Temperature of ions and electrons is changed with each timestep
  - Bracketing: Two calculations run at steady temperature, one higher and one lower than hugoniot temperature

Isentropes are Key Paths in Shock Experiments

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- Follow paths where $dS = 0$
- Isentropic release occurs after compression
- Modeling of shock transfer between materials requires isentropes and hugoniots
- Provide information on sound speed, Gruneisen gamma and so can help construct EOS models

QMD Calculation of Isentropes

- Total differential for entropy

$$dS = \frac{\partial S}{\partial T} dT + \frac{\partial S}{\partial V} dV$$

- Use Maxwell relations and assume Γ/V is nearly constant to write relation between temperature-volume points along isentrope:

$$T_{i+1} \approx T_i \exp\left\{ -V_i \frac{\partial P}{\partial E_i} \ln \frac{V_{i+1}}{V_i} \right\}$$

- Ramped runs provide pressure and energy derivatives w.r.t. temperature
- Start from hugoniot point and construct isentrope

Reference: Desjarlais, M.P. *Quantum Molecular Dynamics Simulations for Generating Equation of State Data*, **Atomic Processes in Plasmas**, AIP, 2009.

QMD Calculation of Quasi-Isentropes

- Uses Hugoniot relation

$$2\left(E_2 - E_1\right) = \left(P_2 + P_1\right)\left(\upsilon_1 - \upsilon_2\right)$$

but with previous isentrope point as reference state 1.

- Can calculate in batches, assuming Gruneisen gamma remains constant across a few isentrope points

- Recalculate gamma at hugoniot temperature of last run in batch

- Approximation to ramped QMD isentropes, should be equal in limit of small steps

6 Mbar Release Isentrope Calculations

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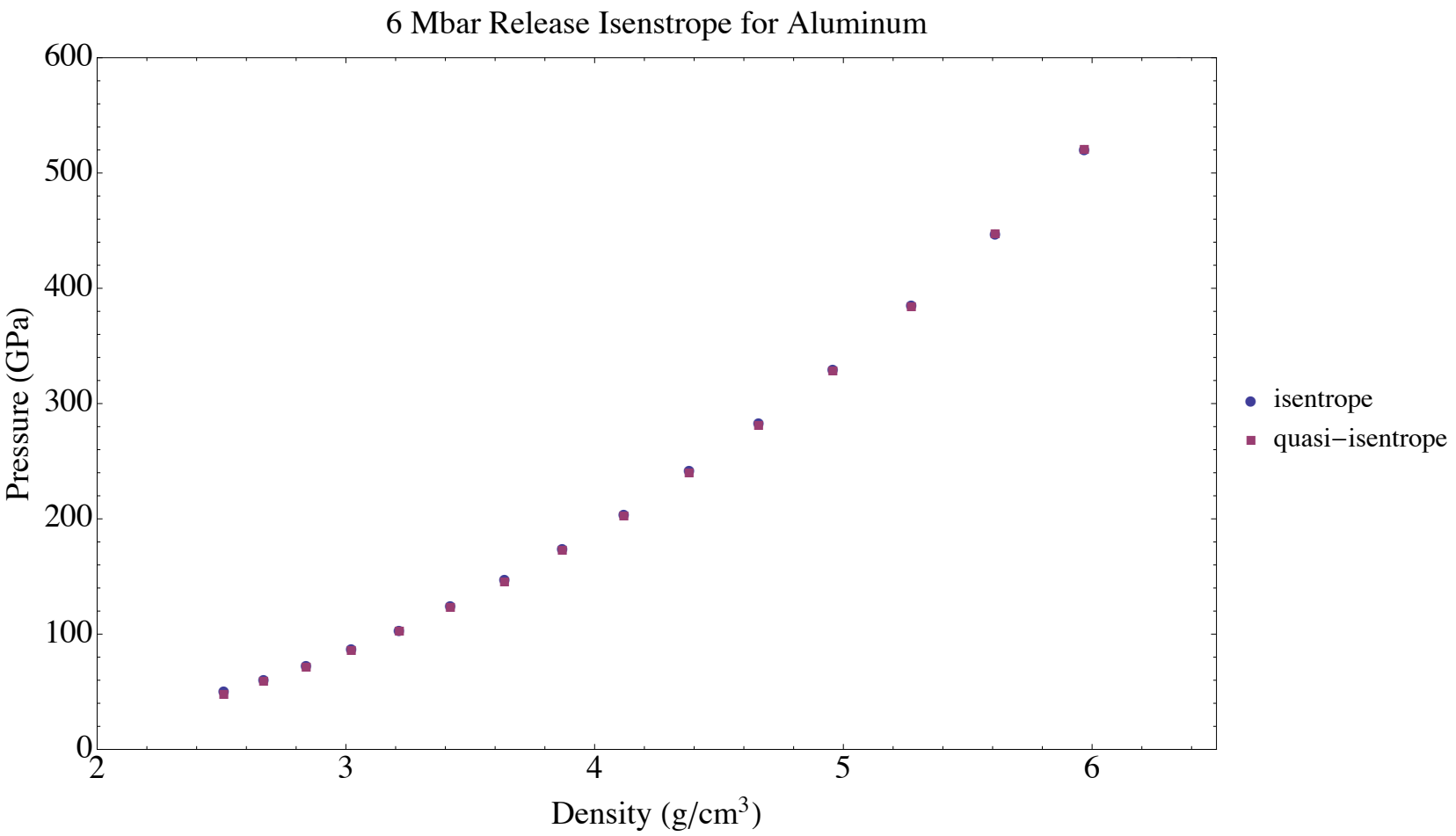
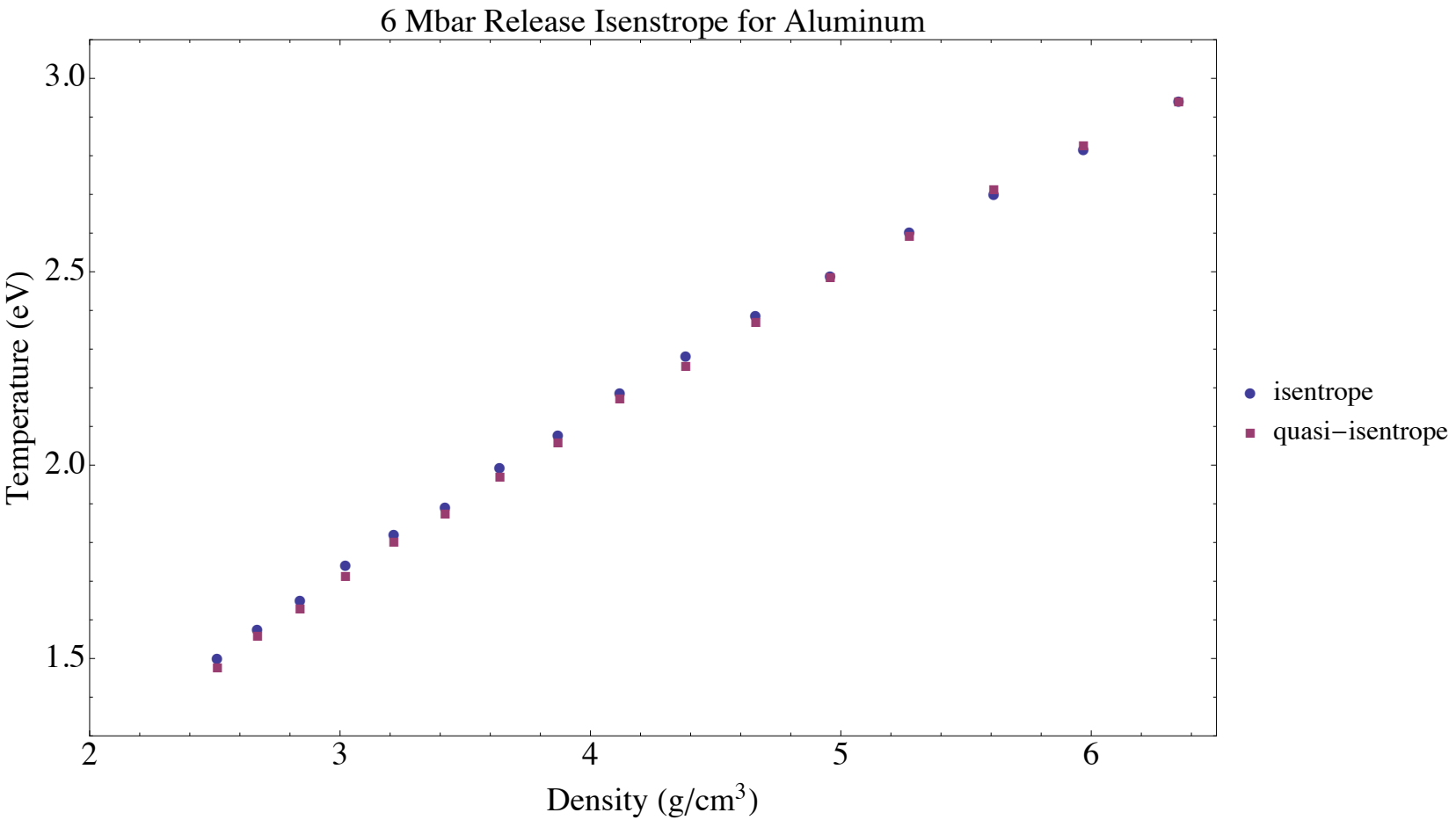
- Aluminum: FCC, 108 atoms
- 3-electron pseudopotential designed by Ann Mattsson
- K-Point Scheme: Baldereschi Mean Value Point
- Algorithm: combined Blocked-Davidson and Reduced Residual Minimization
- Timesteps: between 0.25 and 1 fs
- Ramps: 1 K per timestep

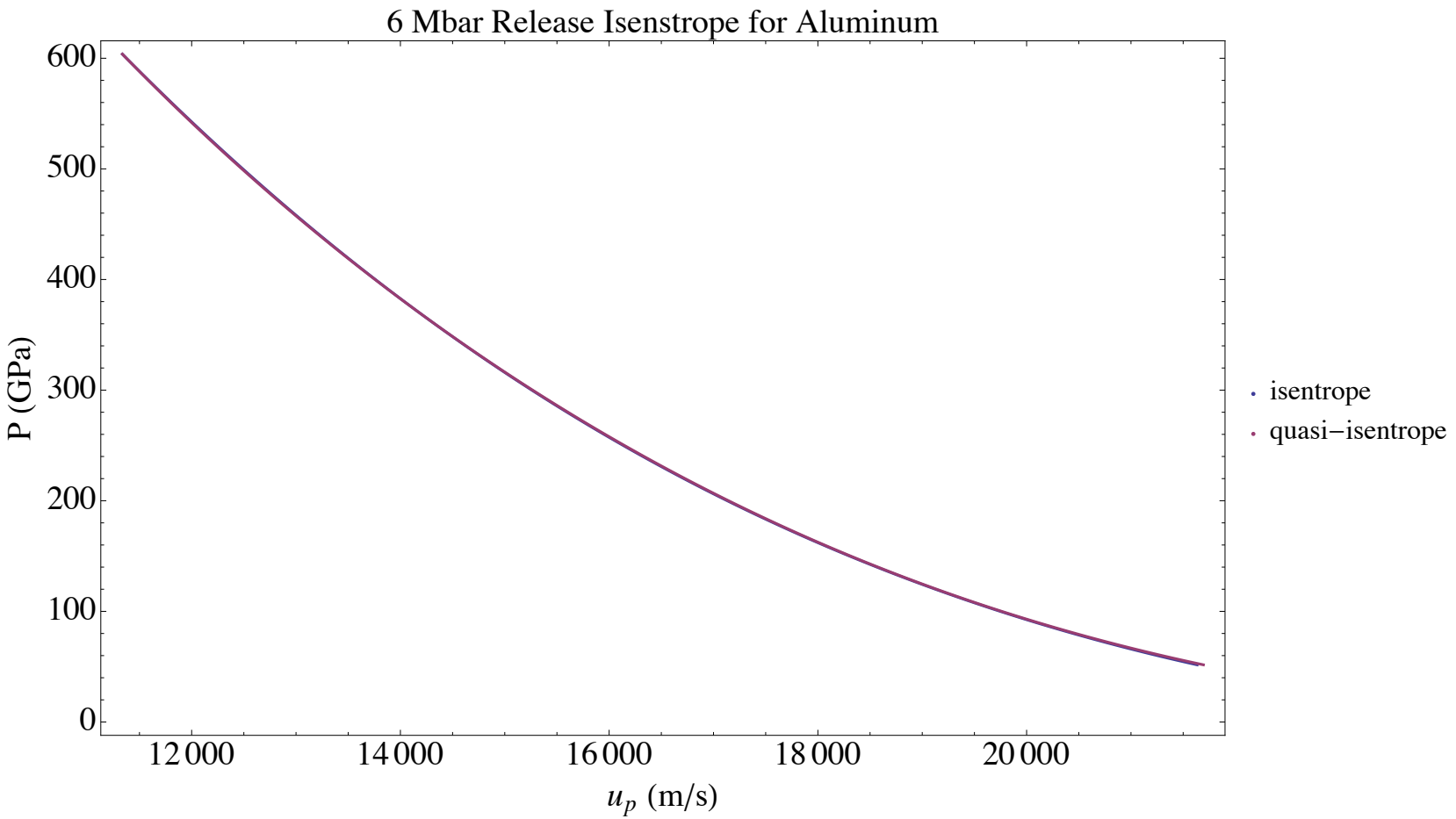
Pressure versus Density



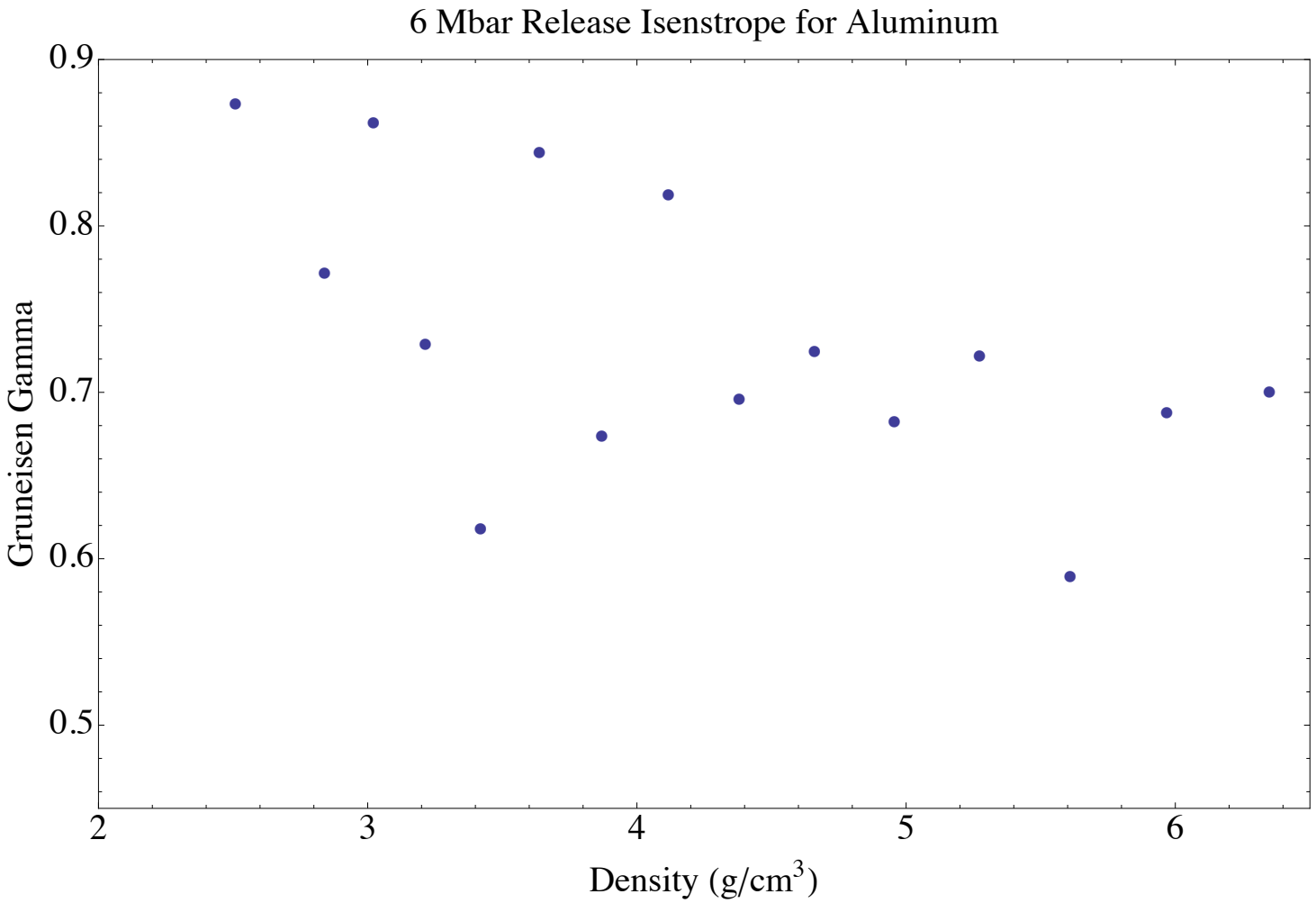
Temperature versus Density



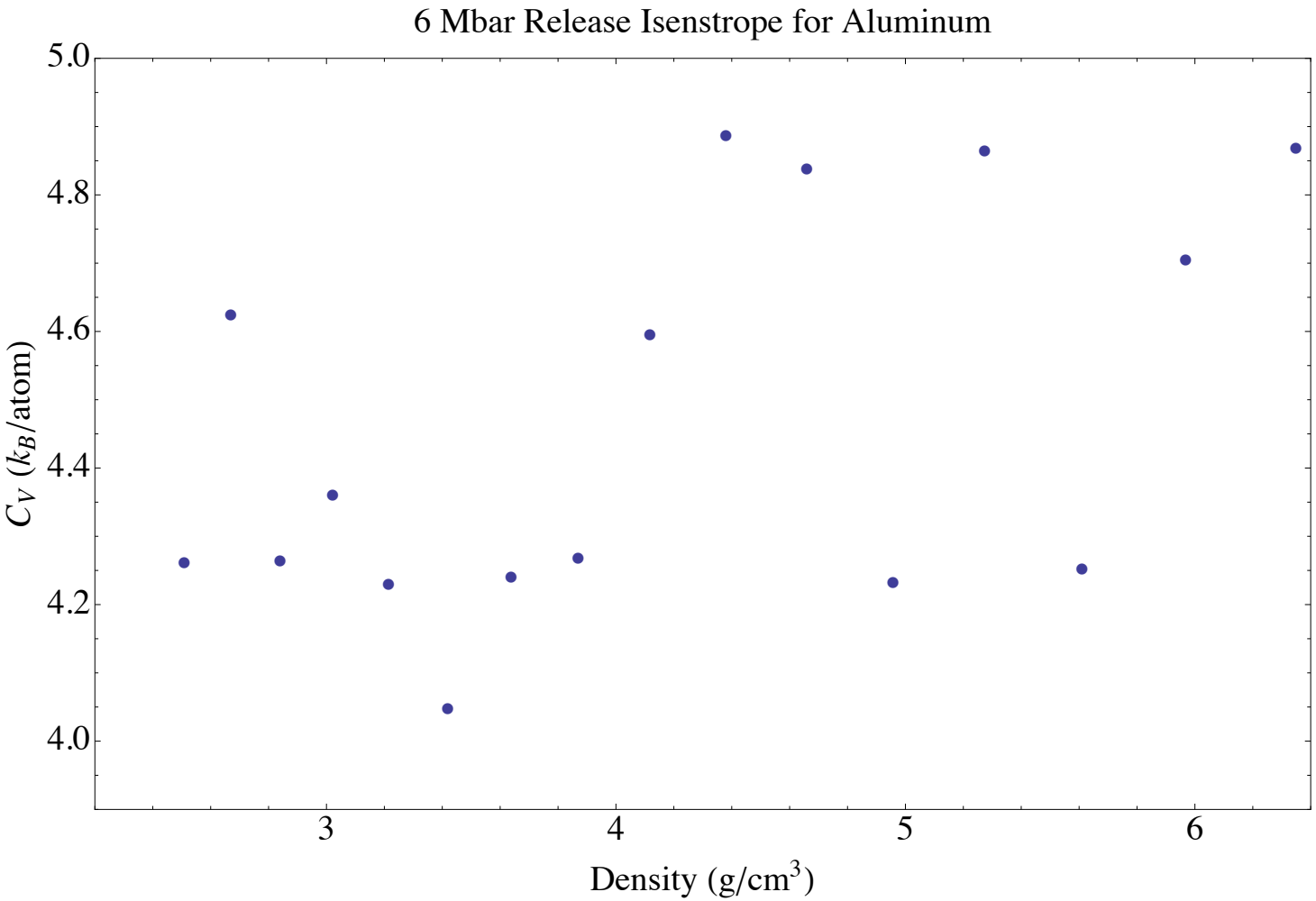
Pressure versus Particle Velocity



Differential Quantities: Gruneisen Gamma



Differential Quantities: Specific Heat



Future Work

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- Continue 6Mbar isentrope calculations
- 12Mbar isentrope with 11-electron pseudopotential
- Compare to experimental data
- Compare 3-electron pseudopotential results to 11-electron pseudopotential results: where do explicit inner electrons become necessary?

Summary

- QMD isentropes and quasi-isentropes give similar results for 6Mbar release isentrope of aluminum, to within
  - 60 m/s for particle velocities (<0.3%)
  - 0.03 eV for isentrope temperatures (<<1%)
  - 3 GPa for isentrope pressures (<4%)
- Close agreement supports use of isentrope method with larger steps than quasi-isentrope method, which requires small hugoniot steps.

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- Kyle Cochrane
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